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Binary mixtures of 1-butyl-1-methylpyrrolidinium bis[(trifluoromethyl)sulfonyl]imide and aliphatic nitrile solvents as electrolyte for EDLCs

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Electrochemical double layer capacitors (EDLCs), also known as supercapacitors, are promising energy storage devices, especially when considering high power applications [1]. EDLCs can be charged and discharged within seconds [1], feature high power ($10 \text{ kW} \cdot \text{kg}^{-1}$) and an excellent cycle life ($>500,000$ cycles). All these properties are a result of the energy storage process of EDLCs, which relies on storing energy by charge separation instead of chemical redox reactions, as utilized in battery systems. Upon charging, double layers are forming at the electrode/electrolyte interface consisting of the electrolyte's ions and electric charges at the electrode surface.

In state-of-the-art EDLC systems activated carbons (AC) are used as active materials and tetraethylammonium tetrafluoroborate ($[\text{Et}_4\text{N}][\text{BF}_4]$) dissolved in organic solvents like propylene carbonate (PC) or acetonitrile (ACN) are commonly used as the electrolyte [2]. These combinations of materials allow operative voltages up to 2.7 V - 2.8 V and an energy in the order of $5 \text{ Wh} \cdot \text{kg}^{-1}$ [3]. The energy of EDLCs is dependent on the square of the operative voltage, thus increasing the usable operative voltage has a strong effect on the delivered energy of the device [1]. Due to their high electrochemical stability, ionic liquids (ILs) were thoroughly investigated as electrolytes for EDLCs, as well as, batteries, enabling high operating voltages as high as 3.2 V - 3.5 V for the former [2]. While their unique ionic structure allows the usage of neat ILs as electrolyte in EDLCs, ILs suffer from low conductivity and high viscosity increasing the intrinsic resistance and, as a result, a lower power output of the device. In order to overcome this issue, the usage of blends of ionic liquids and organic solvents has been considered a feasible strategy as they combine high usable voltages, while still retaining good transport properties at the same time.

In our recent work the ionic liquid N-butyl-N-methylpyrrolidinium bis[(trifluoromethyl)sulfonyl]imide ($[\text{Pyrr}_{14}][\text{TFSI}]$) was combined with two nitrile-based organic solvents, namely butyronitrile (BTN) and adiponitrile (ADN), and the resulting blends were investigated regarding their usage in electrochemical double layer capacitors [4,5]. Firstly, the physicochemical properties were investigated, showing good transport properties for both blends, which are similar to the state-of-the-art combination of $[\text{Et}_4\text{N}][\text{BF}_4]$ in PC. Secondly, the electrochemical properties for EDLC application were studied in depth revealing a high electrochemical stability with a maximum operative voltage as high as 3.7 V. In full cells these high voltage organic solvent based electrolytes have a good performance in terms of capacitance and an acceptable equivalent series resistance at cut-off voltages of 3.2 and 3.5 V. However, long term stability tests by float testing revealed stability issues when using a maximum voltage of 3.5 V for prolonged time, whereas at 3.2 V no such issues are observed.

Considering the obtained results, the usage of ADN and BTN blends with $[\text{Pyrr}_{14}][\text{TFSI}]$ in EDLCs appears to be an interesting alternative to state-of-the-art organic solvent based electrolytes, allowing the usage of higher maximum operative voltages while having similar transport properties to $1 \text{ mol} \cdot \text{dm}^{-3}$ $[\text{Et}_4\text{N}][\text{BF}_4]$ in PC at the same time.

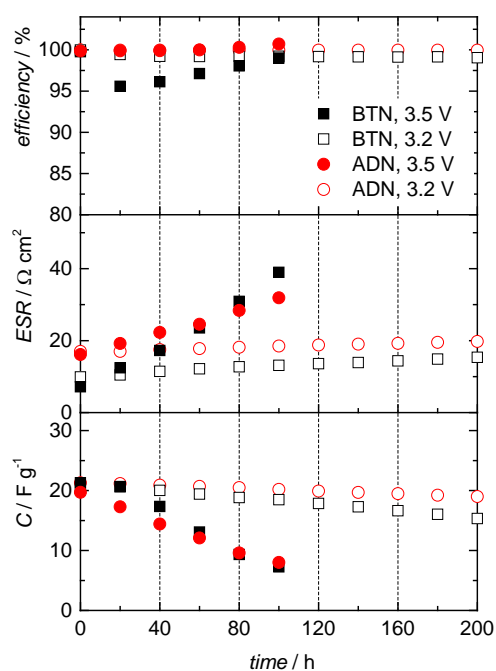


Fig. 1: Evolution of specific capacitance, ESR and efficiency for 200 h float test using blends of butyronitril (BTN)/[Pyrr₁₄][TFSI] and adiponitrile (ADN)/[Pyrr₁₄][TFSI], respectively.

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